UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Ex parte LI YANG and TOSHIHIRO YOSHIDA[1]

Appeal 2007-1524
Application 09/770,725
Technology Center 1700

Decided: June 14, 2007

Before ADRIENE LEPIANE HANLON, ROMULO H. DELMENDO, and SALLY G. LANE, *Administrative Patent Judges*.

DELMENDO, *Administrative Patent Judge*.

DECISION ON APPEAL

STATEMENT OF THE CASE

Applicants appeal under 35 U.S.C. § 134(a) (2005) from a rejection of claims 1-17, which are all the claims pending in the subject application. (Answer entered January 19, 2006; Substitute Answer entered August 22, 2006.) We have jurisdiction under 35 U.S.C. § 6(b) (2005).

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[1] The real party in interest is NGK Insulators, Ltd. (Appeal Br. 1).

Applicants state that they have invented "a lithium secondary battery in which deterioration of battery properties attributed to water released from both of a positive electrode and negative electrode and existing in the non-aqueous electrolytic solution packed in the battery its [sic] suppressed." (Original Specification 1:5-10).

Representative claim 1 reads as follows:

> 1. A lithium secondary battery comprising:
> an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator, said positive electrode comprising positive electrode active substance comprising lithium manganese oxide, said negative electrode comprising negative electrode active substance comprising at least one material selected from the group consisting of amorphous carbonaceous material and graphitized carbonaceous material, and
> a non-aqueous electrolytic solution containing a lithium compound as an electrolyte, wherein a cumulative concentration of water (H_2O) released from both of said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of weight of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C.

The Examiner rejected claims 1-17 under 35 U.S.C. § 103(a). (Substitute Answer entered August 22, 2006, hereafter "Answer" 3-5.)

The prior art relied upon by the Examiner in rejecting the claims on appeal is:

Watanabe	US 6,083,644 B1	Jul. 4, 2000
Takami	US 6,350,544 B1	Feb. 26, 2002
Kurose	US 6,361,822 B1	Mar. 26, 2002

The Examiner's position is that although Takami does not teach the cumulative concentration of water released from the electrodes when subjected to the specified conditions as recited in appealed claim 1, the prior art teachings as a whole would have led one of ordinary skill in the art to reduce the amount of the moisture in the electrodes in order to avoid the known problems associated with moisture. (Answer 4-5.)

Applicants, on the other hand, contend that the claimed subject matter would not have been obvious to a person having ordinary skill in the art over the combined teachings of Takami, Watanabe, and Kurose because Watanabe and Kurose disclose electrode materials different from those described in Takami and thus their teachings with respect to avoidance of moisture have not been shown to be applicable to Takami's batteries. (Appeal Brief filed November 14, 2005, hereinafter "Appeal Br.," at 12; First Reply Brief filed on February 21, 2006, hereinafter "Reply Br. 1," at 2-4; Second Reply Brief filed October 19, 2006, hereinafter "Reply Br. 2," at 2-3, 5-6.) Applicants further contend that while Kurose teaches lowering the water content in the positive electrode material to avoid a decrease in battery charge/discharge capacity, an increase in internal resistance, and deterioration of preservation property, the reference does not quantify the amount of moisture that would be considered detrimental. (*See, e.g.*, Reply Br. 2 at 4.) Applicants also urge that Watanabe discloses drying at a temperature preferably in the range of 80 to 350°C to eliminate the moisture and then assembling the battery but that heating to more than 200°C "is not realistic" because the "binder contained in the electrode would normally be decomposed or would deteriorate at such temperatures." (Appeal Br. 12-13.)

We affirm.

ISSUE

Have Applicants shown that the Examiner erred in concluding that one of ordinary skill in the art would have found it obvious to reduce the amount of moisture in Takami's battery to the levels recited in appealed claim 1 in view of Watanabe and Kurose, thus arriving at a battery encompassed by appealed claim 1?

FINDINGS OF FACT

1. Applicants' Specification states that the lithium manganese oxide material may contain one or more other elements including Si and Ni. (Specification 9:7-27.)
2. Thus, the term "lithium manganese oxide" recited in appealed claim 1 reads on a Ni- and/or Si-containing manganese oxide.
3. Takami discloses a lithium secondary battery comprising a positive electrode, a negative electrode comprising a carbonaceous material capable of absorbing and desorbing Li ions, and a non-aqueous electrolyte, wherein the carbonaceous material has a region of amorphous carbon structure and a region of graphite structure and has a true density of 1.8 g/cm^3 or more and a peak in powder X-ray diffraction corresponding to not more than 0.340 nm in an interplanar spacing d_{002} derived from (002) reflection. (Takami, 2:55-65.)
4. Takami also describes a cylindrical non-aqueous secondary battery in which an electrode assembly **3** is constructed such

that a strip-like laminate body comprising a positive electrode **4**, a separator **5**, and a negative electrode **6** stacked in this order is spirally wound with the separator being disposed at the outermost side of the electrode assembly **3**. (Takami, 4:7-15; Figure 1.)

5. Takami teaches that the positive electrode may be made from "various kinds of oxides," thus indicating to one skilled in the relevant art that the oxides for use as the positive electrode material are not particularly limited. (Takami, 4:39-46.)

6. Nevertheless, Takami states that lithium cobalt oxide, lithium nickel oxide, and lithium manganese oxide are preferred. (Takami, 4:46-49.)

7. Takami further teaches that the carbonaceous material of the negative electrode should be a multi-phase structure having a region of graphite structure and a region of amorphous carbon structure. (Takami, 5:2-7.)

8. As to the non-aqueous electrolyte, Takami discloses that a lithium salt such as lithium hexafluorophosphate ($LiPF_6$), among other possibilities, may be incorporated. (Takami, 10:41-48.)

9. That Takami describes a "positive electrode active substance comprising lithium manganese oxide," a "negative electrode active substance comprising at least one material selected from the group consisting of amorphous carbonaceous material and graphitized carbonaceous material," "a non-aqueous electrolytic solution containing a lithium compound as an electrolyte," and

"an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator," all as recited in appealed claim 1, is not contested.

10. Compared to the subject matter of appealed claim 1, Takami does not disclose "a cumulative concentration of water (H_2O) released from both of the said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of weight of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C."

11. Takami discloses that water is one of the main impurities in the electrolyte solvent that causes "the formation of an insulating film on the surface of a graphitized substance, thereby increasing the surface resistance of the electrodes." (Takami, 10:25-29.)

12. Takami discloses that the formation of the insulating film on the surface of the graphitized substance as a result of water impurities "may give a bad influence to the battery, thereby not only deteriorating the cycle life or capacity thereof, but also increasing the-self-discharge during a high temperature (60° C. or more) storage of the battery." (Takami, 10:30-35.)

13. Accordingly, Takami discloses that it would be desirable to eliminate water as an impurity as much as possible, such that the water content in the electrolyte does not exceed 50 ppm. (Takami, 10:38-39.)

14. Watanabe discloses a non-aqueous electrolyte secondary battery comprising a lithium-containing silicon oxide electrode as a negative electrode, either a lithium-containing titanium oxide or lithium-containing iron sulfide as a positive electrode, and a non-aqueous electrolyte. (Watanabe, 3:45-50.)

15. Watanabe teaches that the negative electrode may contain non-metals such as carbon in addition to the lithium-containing silicon oxide and specifically discusses the use of calcined carbonaceous compounds capable of absorbing and releasing lithium ion or lithium metal. (Watanabe, 6:2-14, 7:37-40.)

16. Watanabe also teaches that the positive electrode material may contain other metals such as nickel and manganese in addition to lithium and titanium. (Watanabe, 8:22-34.)

17. Watanabe discloses the use of lithium salts such as lithium phosphorus hexafluoride ($LiPF_6$) as the electrolyte. (Watanabe, 12:44-46.)

18. Watanabe discloses the desirability of the battery to be assembled in a moisture-free atmosphere. (Watanabe, 14:37-38.)

19. Watanabe discloses the moisture content to be preferably 2,000 ppm or less for the entire battery and 50 ppm for the positive electrode mixture, the negative electrode mixture, or the electrolyte from the point of cycle property. (Watanabe, 14:47-51.)

20. Kurose discloses a non-aqueous electrolyte battery including a nickel-containing lithium composite oxide as an electrode active material. (Kurose, 2:35-40.)

21. Kurose's nickel-containing lithium composite oxide has the formula $Li_xNi_yM_zO_2$ (where x satisfies $0.8<x<1.5$, y+z satisfies $0.8<y+z<1.2$, z satisfies $0\leq z<0.35$, and M is at least one element selected from Co, Mg, Ca, Sr, Al, Mn, and Fe). (Kurose, 2:35-43.)

22. In the working examples, Kurose teaches the use of the nickel-containing lithium composite oxide as a positive electrode material, metallic lithium as a negative electrode, and a non-aqueous electrolytic solution containing $LiPF_6$. (Kurose, 8:42-67.)

23. Kurose teaches (2:14-18):

> Use of an active material in a state with a lot of absorbed moisture in battery causes problems such as a decrease in a charge/discharge capacity of the battery, increase in internal resistance, and deterioration of the preservation property.

24. Applicants rely on the 37 CFR § 1.132 Declaration of Toshihiro Yoshida, one of the named inventors, to demonstrate that "nickel-containing lithium composite oxides do not behave in a manner anywhere near equivalent to $LiMn_2O_4$." (Reply Br. 1 at 4.; Declaration under 37 CFR § 1.132 filed on April 8, 2005.)

25. The Yoshida Declaration states the purpose of the experiments discussed therein as "to show the effect of HF in electrolyte solutions upon dissolution of transition-metal contained in

lithium transition metal compound oxide, and to clarify the difference in the respective amounts of transition metal dissolved into electrolyte solution between lithium nickel oxide and lithium manganese oxide." (Declaration 2.)

26. Applicants do not rely on any evidence to demonstrate that variations in the composition of the lithium oxide positive electrode material significantly affect absorption and release of water (i.e., that the variations in the composition of the lithium oxide positive electrode material are critical to whether water detrimentally affects the characteristics of the battery).

27. Applicants do not assert, much less rely on evidence to show, that one of ordinary skill in the art would not have expected that moisture would cause problems in the type of battery described in Takami.

PRINCIPLES OF LAW

The factual inquiry into whether claimed subject matter would have been obvious includes a determination of: (1) the scope and content of the prior art; (2) the differences between the claimed subject matter and the prior art; (3) the level of ordinary skill in the art; and (4) secondary consideration (e.g., the problem solved) that may be indicia of (non)obviousness. *Graham v. John Deere Co. of Kansas City*, 383 U.S. 1, 17-18 (1966). The Supreme Court of the United States has stated that "[t]he obviousness analysis cannot be confined by a formalistic conception of the words teaching, suggestion, and motivation, or by overemphasis on the importance of published articles and the explicit content of issued patents." *KSR Int'l Co. v. Teleflex, Inc.*,

127 S. Ct. 1727, 1741, 82 USPQ2d 1385, 1396, (2007). Rather, "[w]hen there is a design need or market pressure to solve a problem and there are a finite number of identified, predictable solutions, a person of ordinary skill in the art has good reason to pursue the known options within his or her technical grasp." *KSR*, 127 S. Ct. at 1732, 82 USPQ2d at 1397.

"When the PTO shows prima facie obviousness, the burden then shifts to the applicant[s] to rebut." *In re Mayne*, 104 F.3d 1339, 1342, 41 USPQ2d 1451, 1454 (Fed. Cir. 1997). "Such rebuttal or argument can consist of a comparison of test data showing that the claimed compositions possess unexpectedly improved properties or properties that the prior art does not have..." *In re Dillon*, 919 F.2d 688, 692-93, 16 USPQ2d 1897, 1901 (Fed. Cir. 1990)(en banc).

ANALYSIS

Applicants have argued claims 1-17 together. We select claim 1 as representative of all the appealed claims 1-17. We therefore confine our discussion to this representative claim. Furthermore, any argument not made has been waived. 37 C.F.R. § 41.37(c)(vii).

Turning to the merits, Takami discloses a lithium secondary battery comprising a positive electrode, a negative electrode comprising a carbonaceous material capable of absorbing and desorbing Li ions, and a non-aqueous electrolyte, wherein the carbonaceous material has a region of amorphous carbon structure and a region of graphite structure and has a true density of 1.8 g/cm^3 or more and a peak in powder X-ray diffraction corresponding to not more than 0.340 nm in an interplanar spacing d_{002} derived from (002) reflection. (Takami, 2:55-65.) In particular, Takami

describes a cylindrical non-aqueous secondary battery in which an electrode assembly **3** is constructed such that a strip-like laminate body comprising a positive electrode **4**, a separator **5**, and a negative electrode **6** stacked in this order is spirally wound with the separator being disposed at the outermost side of the electrode assembly **3**. (Takami, 4:7-15; fig. 1.) Takami further teaches that the positive electrode may be made from "various kinds of oxides," thus indicating to one skilled in the relevant art that the oxides for use as the positive electrode material are not particularly limited, but states that lithium cobalt oxide, lithium nickel oxide, and lithium manganese oxide are preferred. (Takami, 4:39-49.) Takami also teaches that the carbonaceous material of the negative electrode should be a multi-phase structure having a region of graphite structure and a region of amorphous carbon structure. (Takami, 5:2-7.) As to the non-aqueous electrolyte, Takami discloses that a lithium salt such as lithium hexafluorophosphate ($LiPF_6$), among other possibilities, may be incorporated. (Takami, 10:41-48.)

That Takami describes a "positive electrode active substance comprising lithium manganese oxide," a "negative electrode active substance comprising at least one material selected from the group consisting of amorphous carbonaceous material and graphitized carbonaceous material," "a non-aqueous electrolytic solution containing a lithium compound as an electrolyte," and "an electrode unit produced by winding or laminating a positive electrode and a negative electrode via a separator," all as recited in appealed claim 1, is not contested.

Watanabe discloses a non-aqueous electrolyte secondary battery comprising a lithium-containing silicon oxide electrode as a negative

electrode, either a lithium-containing titanium oxide or lithium-containing iron sulfide as a positive electrode, and a non-aqueous electrolyte. (Watanabe, 3:45-50.) Watanabe teaches that the negative electrode may contain non-metals such as carbon in addition to the lithium-containing silicon oxide and specifically discusses the use of calcined carbonaceous compounds capable of absorbing and releasing lithium ion or lithium metal. (Watanabe, 6:2-14, 7:37-40.) As to the positive electrode, Watanabe teaches that it may contain other metals such as nickel and manganese in addition to lithium and titanium. (Watanabe, 8:22-34.) Watanabe also discloses the use of lithium salts such as lithium phosphorus hexafluoride ($LiPF_6$) as the electrolyte. (Watanabe, 12:44-46.)

According to Watanabe, the battery is desirably assembled in a moisture-free atmosphere. (Watanabe, 14:37-38.) Specifically, Watanabe discloses the moisture content to be preferably 2,000 ppm or less for the entire battery and 50 ppm for the positive electrode mixture, the negative electrode mixture, or the electrolyte from the point of cycle property. (Watanabe, 14:47-51.)

Kurose discloses a non-aqueous electrolyte battery including a nickel-containing lithium composite oxide as an electrode active material. (Kurose, 2:35-40.) Kurose's nickel-containing lithium composite oxide has the formula $Li_xNi_yM_zO_2$ (where x satisfies $0.8<x<1.5$, y+z satisfies $0.8<y+z<1.2$, z satisfies $0\leq z<0.35$, and M is at least one element selected from Co, Mg, Ca, Sr, Al, Mn, and Fe). (Kurose, 2:35-43.) In the working examples, Kurose teaches the use of the nickel-containing lithium composite oxide as a positive electrode material, metallic lithium as a negative

electrode, and a non-aqueous electrolytic solution containing $LiPF_6$.

(Kurose, 8:42-67.) Kurose teaches (2:14-18):

> Use of an active material in a state with a lot of absorbed moisture in battery causes problems such as decrease in a charge/discharge capacity of the battery, increase in internal resistance, and deterioration of the preservation property.

Thus, Kurose (like Watanabe) explicitly states that moisture in a positive electrode that is compositionally and structurally similar to that described in Takami causes problems in lithium batteries.

Compared to the subject matter of appealed claim 1, Takami does not disclose "a cumulative concentration of water (H_2O) released from both of the said positive electrode and said negative electrode in relation to the weight of said electrode unit, exclusive of current collectors, is suppressed to 5,000 ppm or lower in case of heating both electrodes at 25 to 200°C and to 1,500 ppm or lower in case of heating said electrodes at 200 to 300°C." Nevertheless, we agree with the Examiner's conclusion that the claimed subject matter as a whole would have been obvious to a person of ordinary skill in the art over the prior art.

We start with the negative electrode. Takami discloses that water in the electrolyte solvent is one of the main impurities that causes "the formation of an insulating film on the surface of a graphitized substance, thereby increasing the surface resistance of the electrodes." (Takami, 10:27-30.) This may, in turn, "give a bad influence to the battery, thereby not only deteriorating the cycle life or capacity thereof, but also increasing the-self-discharge during a high temperature (60° C. or more) storage of the battery." (Takami, 10:31-35.) For these reasons, Takami discloses that it would be desirable to eliminate water as an impurity from the electrolyte containing a

non-aqueous solvent as much as possible, such that the content of water does not exceed 50 ppm. (Takami, 10:38-39.)

These teachings in Takami would have led a person having ordinary skill in the art to reasonably draw an inference that the presence of water in the carbonaceous material of the negative electrode should also be avoided. Furthermore, Watanabe teaches that the moisture content of a negative electrode that may contain calcined carbonaceous material (in addition to lithium-containing silicon oxide) in a similar battery should be below 50 ppm "from the point of cycle property." (Watanabe, 7:37-40, 14:37-51.) Under these circumstances, we conclude that one of ordinary skill in the art would have been led to reduce the amount of moisture in Takami's negative electrode to the greatest extent possible, such as 50 ppm or lower, in order to avoid the known problems associated with moisture, as disclosed in both Takami and Watanabe.

With respect to the positive electrode, we have found that Kurose teaches that a positive electrode including nickel-containing lithium oxide, which may further contain Mn, absorbs moisture, causing problems such as a decrease in charge/discharge capacity, an increase in internal resistance, and deterioration of preservation property. In a similar fashion, Watanabe also cautions against the presence of moisture for a lithium-titanium oxide based positive electrode that may additionally contain Mn and Ni. Based on the reasonable expectation that the problems caused by moisture as described in Kurose and Watanabe would also occur in other electrodes of similar composition, we conclude that one of ordinary skill in the art would have found it obvious to reduce the amount of water in Takami's positive

electrode to the greatest extent possible, e.g., 50 ppm as disclosed in Watanabe.

When the moisture contents of Takami's negative and positive electrodes are modified in the manner discussed above, one of ordinary skill in the art would have arrived at a lithium secondary battery encompassed by appealed claim 1. Having determined that the Examiner has established a prima facie case of obviousness, we consider the Applicants' relied upon arguments and evidence in rebuttal.

Applicants urge that the claimed subject matter would not have been obvious to a person having ordinary skill in the art over the combined teachings of Takami, Watanabe, and Kurose because Watanabe and Kurose disclose electrode materials different from those described in Takami and thus their teachings with respect to avoidance of moisture have not been shown to be applicable to Takami's batteries. (Appeal Br. at 12; Reply Br. 1, at 2-4; Reply Br. 2 at 2-3, 5-6.)

We find this contention to be without merit. The lithium oxides described as useful in Kurose (nickel-containing lithium composite oxide having the formula $Li_xNi_yM_zO_2$ where M may be Mn) are structurally similar to those described in Takami ($LiNiO_2$, $LiMn_2O_4$, or $LiMnO_2$). Likewise, Watanabe's lithium-containing titanium oxides, which may additionally contain Mn or Ni, are also similar. Thus, one of ordinary skill in the art would have reasonably predicted or had a reasonable expectation that Takami's lithium oxide would also absorb moisture and suffer from the same or similar problems discussed in Kurose and Watanabe.

From the collective teachings of the prior art, one of ordinary skill in the art would have understood the general problems associated with the

presence of moisture in lithium secondary batteries. The desirability of solving these general problems would have led the person of ordinary skill in the art to make the electrodes as free of moisture as possible.

While Applicants would have us believe that slight differences in terms of composition are critical to whether the battery is susceptible to moisture, the prior art suggests otherwise. Both Kurose and Watanabe undercut Applicants' argument because these references teach the importance of eliminating water for a wide variety of electrode compositions. Indeed, Applicants do not rely on any evidence to demonstrate that variations in the composition of the lithium oxide positive electrode material significantly affect absorption and release of water (i.e., that the variations in the composition of the lithium oxide positive electrode material are critical to whether water detrimentally affects the characteristics of the battery). Here, we find it significant that Applicants do not unequivocally assert, much less prove, that one of ordinary skill in the art would not have expected that moisture would cause problems in the type of battery described in Takami.

We have considered the Yoshida Declaration but find that it is insufficient to overcome the Examiner's rejection. That Declaration merely establishes a difference in dissolution rates of transition metal into the electrolyte solution depending on the composition of the positive electrode. Specifically, the data are said to show that the rate of dissolution of Mn from $LiMn_2O_4$ into the electrolyte is much higher than the rate of dissolution of Ni from $LiNiO_2$. But Applicants have not established the relevance or significance of the difference in the dissolution rates to the question of whether one of ordinary skill in the art would have expected moisture to

cause problems in Takami's electrodes. The Examiner's combination of references is not premised on the notion that Kurose's materials would have transition metal dissolution characteristics identical to those of Takami's materials. Rather, it is based on the finding that one of ordinary skill in the art would have had a reasonable expectation that water would be detrimental to Takami's battery, as disclosed in Kurose and Watanabe, based on their structural similarities. That Mn in $LiMn_2O_4$ has a higher dissolution rate than Ni in $LiNiO_2$ does not negate this expectation. As discussed previously, Applicants have not relied on any evidence indicating that one of ordinary skill in the art would not have expected that moisture would cause problems in the type of battery described in Takami.

Applicants further contend that while Kurose teaches lowering the water content in the positive electrode material to avoid a decrease in battery charge/discharge capacity, an increase in internal resistance, and deterioration of preservation property, the reference does not quantify the amount of moisture that would be considered detrimental. (*See*, *e.g.*, Reply Br. 2 at 4.) This argument is also unpersuasive. Given that it was known in the art that moisture is undesirable, a person of ordinary skill in the art would have reduced the amount of moisture to the greatest extent, subject to cost considerations. Applicants have not shown that such reduced levels of moisture would not have included the amounts indirectly recited in appealed claim 1. Moreover, Watanabe explicitly discloses that the amount of moisture in each of the electrodes should be 50 ppm or less.

Applicants argue that Watanabe discloses drying at a temperature preferably in the range of 80 to 350°C to eliminate the moisture and then assembling the battery but that heating to more than 200°C "is not realistic"

because the "binder contained in the electrode would normally be decomposed or would deteriorate at such temperatures." (Appeal Br. 12-13.) Applicants' argument is based solely on a statement of counsel. The arguments of counsel cannot take the place of evidence in the record. *In re Geisler*, 116 F.3d 1465, 1470, 43 USPQ2d 1362, 1365 (Fed. Cir. 1997) ("An assertion of what seems to follow from common experience is just attorney argument and not the kind of factual evidence that is required to rebut a prima facie case of obviousness.") Because Appellants have not proffered any evidence to this effect, Applicants' argument is of no help. Even if evidence had been made of record in this appeal, Watanabe discloses drying temperatures below 200°C, which presumably would not deteriorate the binder.

CONCLUSIONS OF LAW

On the record before us, Applicants have failed to rebut the prima facie case established by the Examiner that a person of ordinary skill in the art would have found the subject matter of appealed claims 1-17 obvious over the prior art.

We therefore affirm the rejection under 35 U.S.C. § 103(a) of all claims.

No time period for taking any subsequent action in connection with this appeal may be extended under 37 C.F.R. § 1.136(a).

AFFIRMED

Appeal 2007-1524
Application 09/770,725

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